# THE EVALUATION OF CONVERTERS FOR EXOTHERMIC AND ENDOTHERMIC CATALYTIC REACTIONS OCCURRING WITHIN NARROW TEMPERATURE LIMITS

BY GUSTAV WIRTH
TRANSLATED BY R. C. GRASS AND H. J. KANDINER

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# THE EVALUATION OF CONVERTERS FOR EXOTHERMIC AND ENDOTHERMIC CATALYTIC REACTIONS OCCURRING WITHIN NARROW TEMPERATURE LIMITS

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Gustav Wirth[1]

Translated by R. C. Grass[2] and H. J. Kandiner[3]

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# PREFACE

(By R. C. Grass and H. J. Kandiner)

The following paper is a translation of T.O.M. Reel 134, Section I-B, Item 37, dated at Leuna, July 4, 1942. The translation was undertaken because considerable interest exists in American technical circles concerning certain design methods used in Germany. It must be pointed out that in this article the author makes his calculations and mathematical analyses largely in terms of "immeasurables"; for example, temperatures "near" the cooling surface and "average" thermal conductivities of the catalyst bed and the gas stream are employed.

It is the opinion of the translators that the particular methods discussed herein are extreme simplifications of the actual physical problems and that their adequate description of the natural phenomena that really occur is questionable. As examples of these simplifications employed by the author, attention is called to the following:

1. The assumption is made that the heat generation per unit volume of catalyst is constant throughout the reactor. As this quantity depends on the amount of reaction taking place per unit volume, and hence on the temperature of the catalyst bed, and as a temperature variation across the bed is presumed, then the heat release per unit volume must necessarily vary.

2. The assumption is also made that all of the heat generated is carried away by conduction through the catalyst bed in a direction transverse to the main gas flow, and that the gases leave the reaction zone at the same temperature at which they enter. In the opinion of the translators, however, it is more likely that a substantial fraction of the heat generated is carried off by the flowing gases.

In spite of the questionable nature of the calculations and mathematical relationships involved in this paper, it is believed that the information contained herein may prove valuable to workers in the field of catalytic-reactor design in providing an insight into the methods employed by the Germans.



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ABSTRACT

In exothermic and endothermic catalytic reactions, control of the temperature is of utmost importance. Basic calculations are developed for determining catalyst volumes of various types of reactors that give particular temperature distributions, and the individual designs are compared.

## INTRODUCTION

A narrow range of permissible operating temperatures characterizes many catalytic reactions, and yields of the desired product are reduced sharply if operating temperatures fall outside of this optimum range. In order to achieve close temperature control, reactors with internal heat exchange have been developed in which thin layers of catalyst are located between cooling surfaces.

The temperature distribution for an exothermic reaction in such a thin catalyst layer is shown in figure 1. Reactive gases flow downward through the catalyst section, which is $2s_o$ wide. It is assumed that a constant amount of heat is generated per unit of bulk catalyst volume, and that this heat is entirely removed through the walls. Consequently, except for a short section at the entrance of the reactor, the heat of reaction is not available for raising the temperature of the gases. Thus, at all points equidistant from the axis, the temperature is the same throughout the entire reactor. At the middle of the gas stream, the temperature is designated as $u_o$. Traveling radially from this point, the temperature decreases because of the removal of heat through the walls and reaches, in the vicinity of the cooling surface, the value $u_1$. Over a relatively narrow zone of width, b, the temperature falls to the value $u_2$ at the cooling surface. Because inadequate data are available on heat-transfer coefficients for a gas flowing through a porous bed, and also because the narrow zone, b, is probably without effect on the reaction by virtue of its low temperature, the temperature drop in this zone at the cooling wall was disregarded. The following calculations were made on the basis of the temperature drop $(u_o - u_1)$ resulting from heat conduction within the main body of the catalyst bed only.

The above considerations supplied a basis for calculations that aided in solution of the following problems:

1. Design of a reactor from physical data; for example, either the thermal conductivity of the catalyst or experimental measurements of the heat released in and the temperature drop across an arbitrary bed.

2. Scaling-up data for various tube shapes from laboratory determination of the maximum permissible dimensions of the catalyst tube.

3. Relation of the cost and efficiency of heat-transfer surfaces for various reactor designs for exothermic and endothermic reactions.



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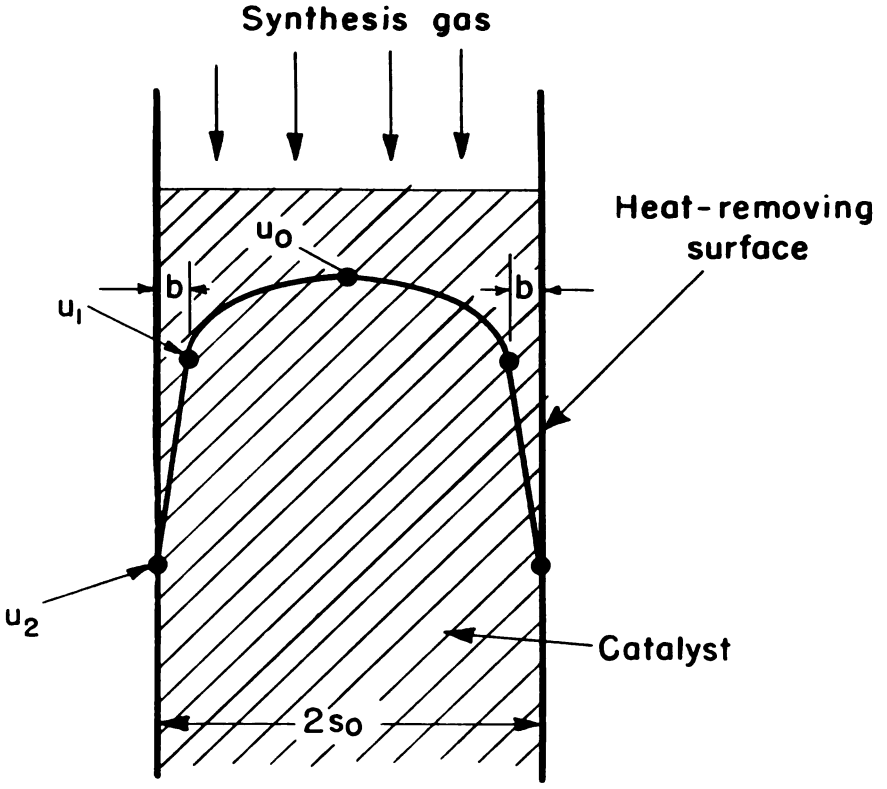


Figure 1. - Temperature distribution in a catalyst-filled section.

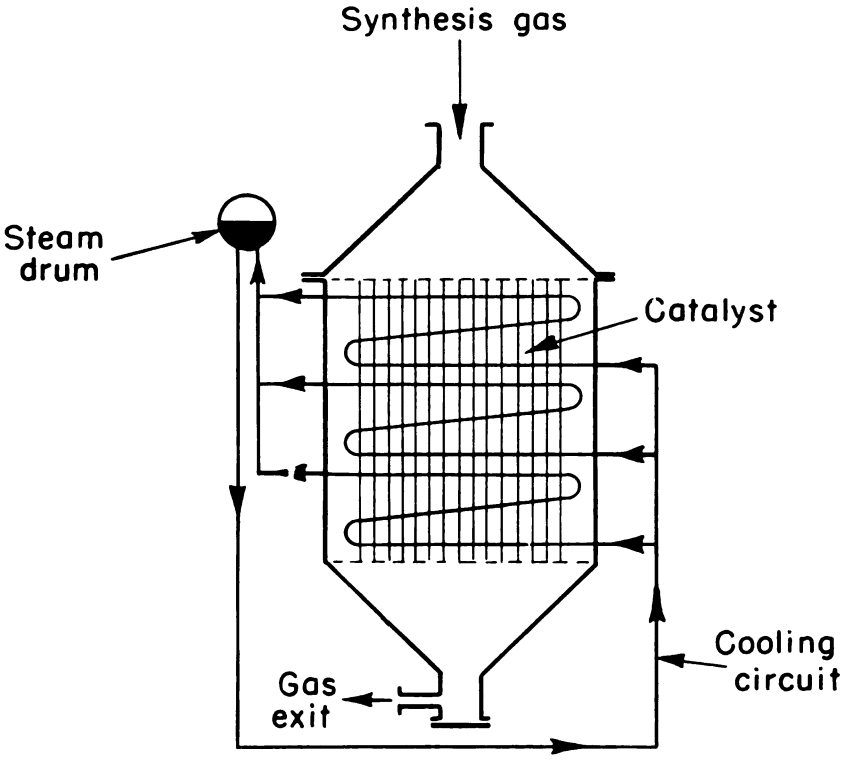


Figure 2. - Plate converter.

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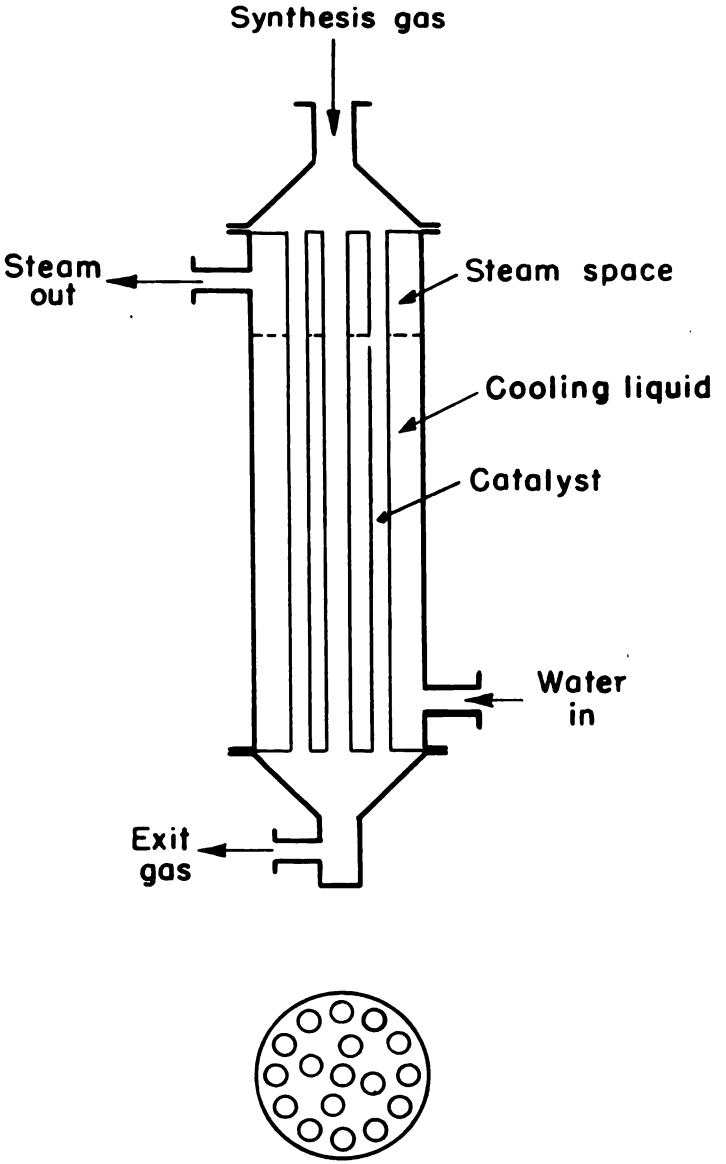


Figure 3. - Inner-catalyst converter.

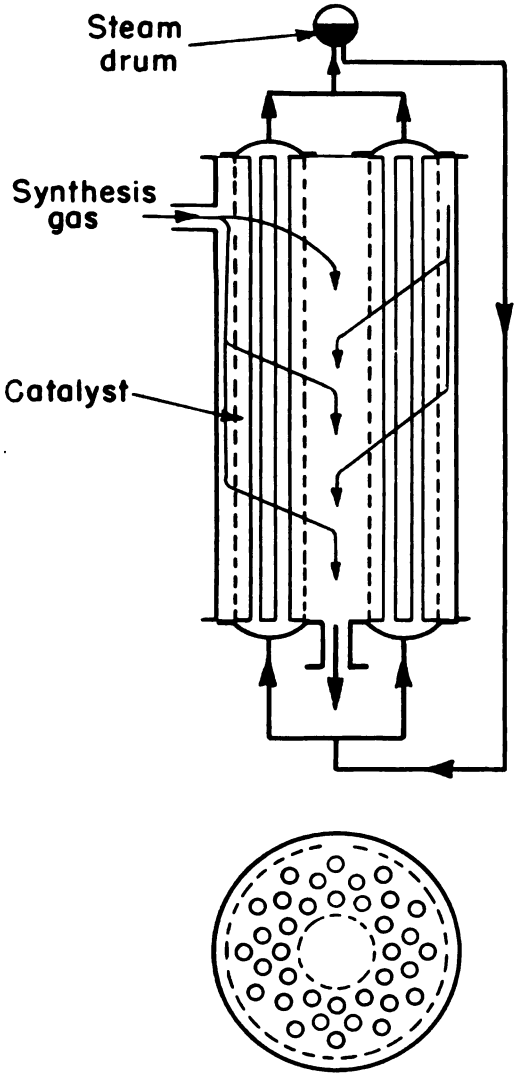


Figure 4. - Outer-catalyst converter.

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Tubular reactors, in which the catalyst is contained in cylindrical tubes, are usually used in the laboratory. For such a converter, the optimum dimensions can easily be obtained by carrying out the reaction in tubes of various diameters. This type of converter, hereafter designated as the "inner-catalyst" converter, is regarded as a standard shape, and other reactor designs are compared to it.

## COMMON REACTOR TYPES

### 1. - Plate Converters

A converter was developed for the Fischer-Tropsch synthesis in which the catalyst was placed between flat cooling surfaces (fig. 2). This converter is characterized by cooling partitions through which tubes carrying a cooling liquid are fastened by peening or rolling. The converter may also be regarded as a finned-tube in which very large fins are penetrated by a large number of parallel tubes. The flow of heat proceeds from the catalyst to the partitions (fins) and through the partitions to the cooling tube; the temperature gradient in a partition can be disregarded because of its low heat capacity. Water at its boiling point was employed as a cooling medium. Steam and water flow to an overhead drum, from which the steam is withdrawn and into which feed water is introduced. The synthesis gas enters above the catalyst bed and is split up by the cooling partitions into a corresponding number of component streams. The reaction products and unconverted feed gas leave the converter at the bottom.

### 2. - Inner-catalyst Converters

In this construction, a number of tubes, filled with catalyst, are connected in parallel by rolling or welding them into top and bottom tube sheets. A pressure-tight shell confines the evaporating cooling medium to the space surrounding the tubes. The gas flows from top to bottom through the tubes. The converter can, as in case (1), be connected to a separate steam collector, or steam-collecting space may be provided inside the converter housing itself, as shown in figure 3. The portions of the tubes in the steam space should not contain catalyst, as overheating will invariably result. These empty sections serve to preheat the gas to synthesis temperature. This type of converter is, in effect, a parallel array of one-tube converters, each similar to those employed in the laboratory.

### 3. - Outer-catalyst Converters

Alternatively, the cooling medium may be passed through the tubes with the catalyst surrounding them (see fig. 4) obtaining, thereby, the "outer-catalyst" converter. The tubes are arranged vertically, as shown in the illustration. Synthesis gas flows from an annular distributing space in a radial direction through the catalyst bed and is removed from the converter, together with the reaction products, through a circular collecting space in the center of the converter. The cooling system is not substantially different from the designs previously discussed.



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## 4. - Annular-volume Converters

Instead of being located inside the tubes in the manner described under (2), the catalyst can be charged into the annular space between two concentric tubes (see fig. 5). The cooling medium will then be in the inner tube as well as around the outer tube. The outer tubes are welded into top and bottom base plates, and the rest of the converter design is similar to the inner-catalyst converter described under (2).

The four converter designs described above were selected for the present investigation. They were used as a basis for the calculations that follow, because the individual cross sections of the catalyst spaces are simple or can be reduced to a simple basis. Other designs are not common and will not be considered; it should be possible, however, by estimating from the following basic curves and calculations, to obtain for other experimental designs an estimate of the appropriate dimensions. Considerations involving exothermic reactions may, fundamentally, be "inverted" for endothermic reactions with valid results.

## THEORETICAL CONSIDERATIONS

The cross sections of the converters previously described fall into one of the classifications shown in figure 6:

1. Rectangular catalyst-filled cross section (plate converter), in which the heat flows from the middle of the section toward the two boundary cooling surfaces.

2. Circular catalyst-filled cross section (inner-catalyst converter), in which the heat flows radially out to the cooling surface.

3. Annular catalyst-filled cross section (outer-catalyst converter), in which the catalyst is located around the cooling tube.

4. Annular catalyst-filled cross section (annular-volume converter), in which the catalyst is located between two concentric tubes. These four cases are separately discussed in the following.4/

### 1. Plate Converter

Nomenclature (refer to fig. 7)

$s$ = distance from median plane to any point in catalyst bed (m).

$2 s_o$ = distance between cooling surfaces, (m).

$u$ = temperature (°C.); $u_o$ = temperature at central plane;

$u_1$ = temperature at cooling surface (assumed constant).

4/ The mathematical derivations of the original have been modified by the translators in order to make them somewhat more concise.



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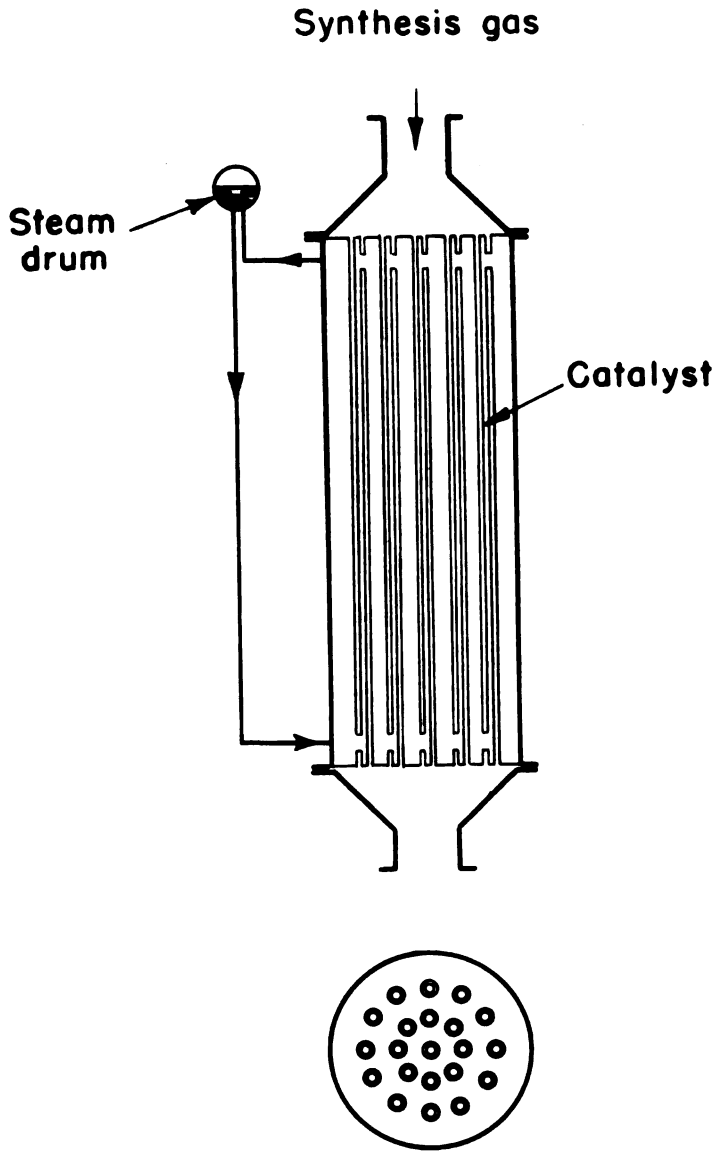


Figure 5. - Annular-volume converter.

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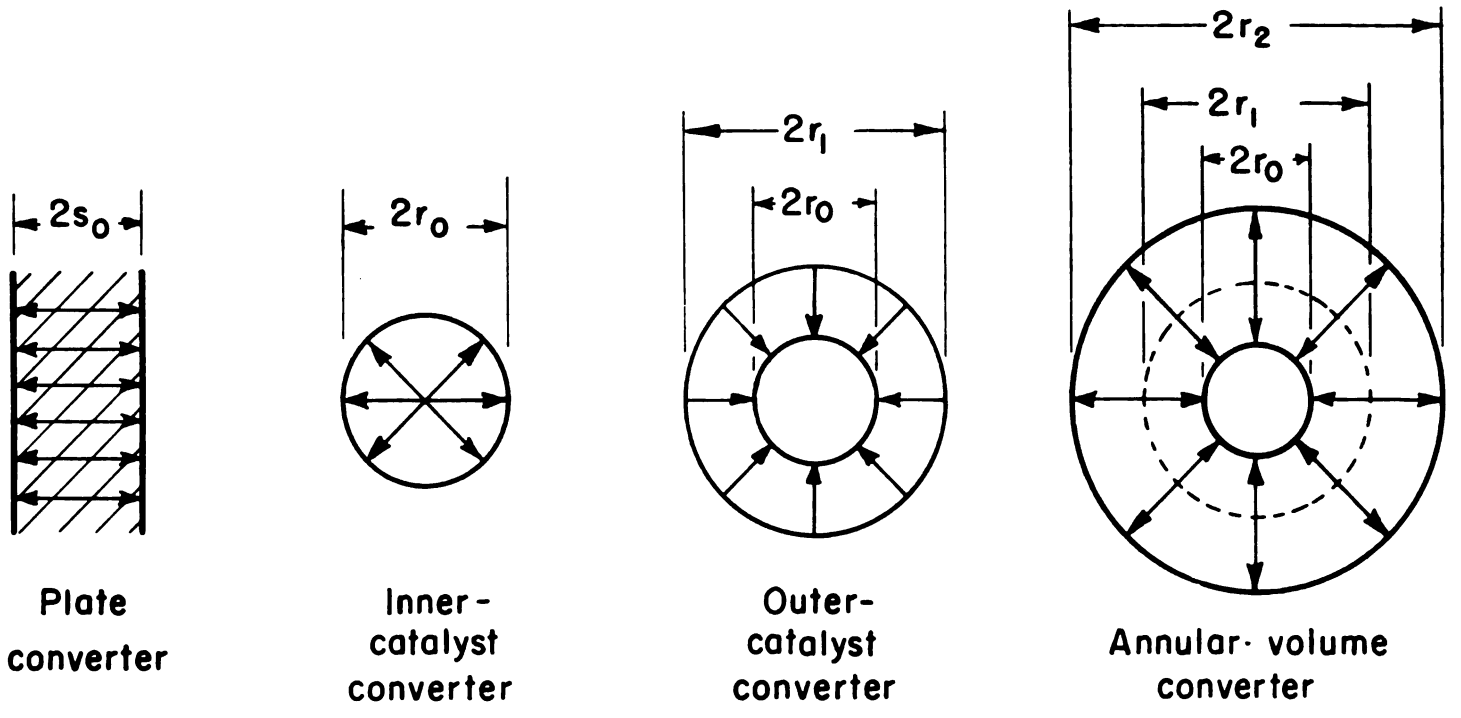


Figure 6. - Types of catalyst-filled cross sections; the direction of heat flow is indicated by arrows.

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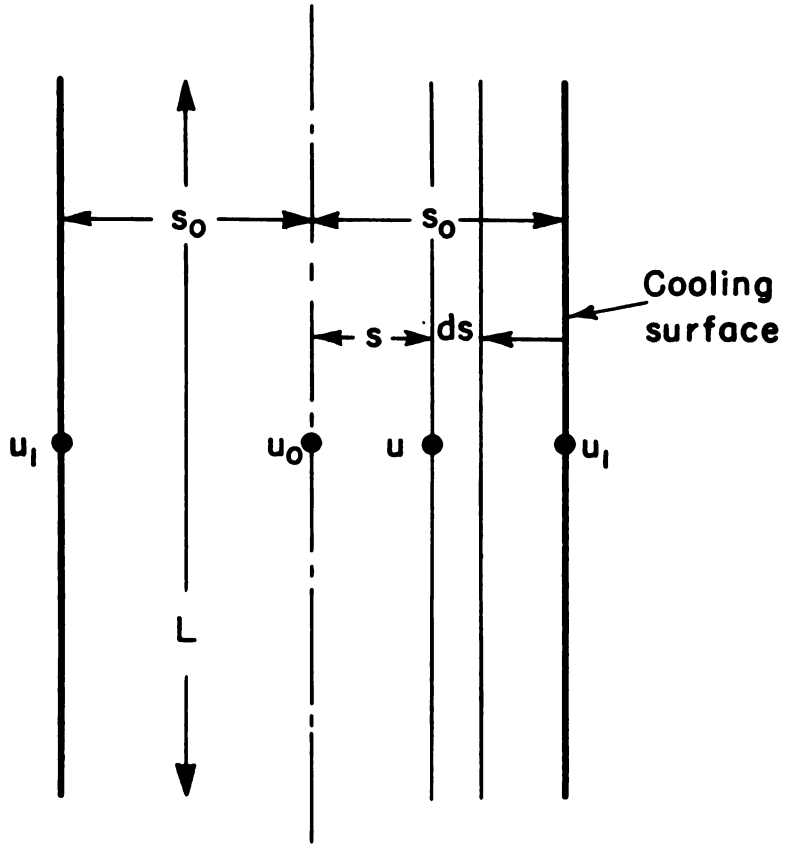


Figure 7. - Catalyst-filled cross section (plate converter).

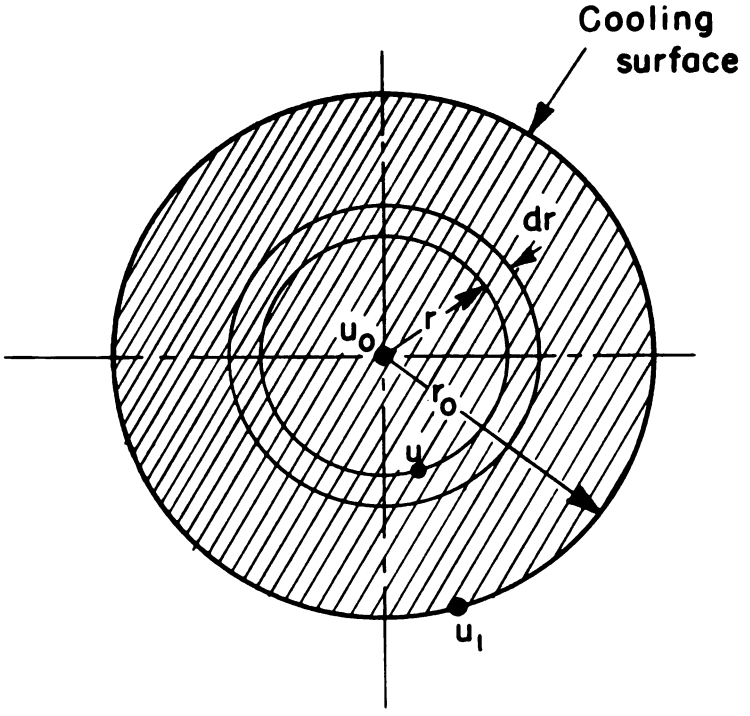


Figure 8. - Cross section of a catalyst-filled tube.

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$\Delta t = u_o - u_1$

$q$ = heat generated in the catalyst bed (kg.-cal./$m^3$ hour).

$\lambda$ = mean thermal conductivity of gas phase and catalyst bed (kg.-cal./m. hour $^oC$.)

Consider an infinite bar $2s_o$ wide and 1 unit thick.

The steady-state temperature distribution in such a bar is then defined by:

$$\lambda \frac{d^2u}{ds^2} + q = 0 \quad \ldots\ldots\ldots\ldots\ldots \quad (1)$$

subject to certain boundary conditions.

By integrating (1), equation (2) is obtained.

$$\frac{du}{ds} = \frac{-qs}{\lambda} + C_1 \quad \ldots\ldots\ldots\ldots\ldots \quad (2)$$

However, at $s = 0$, $\frac{du}{ds} = 0$ (i.e., no heat flows across the midplane of the section), hence $C_1 = 0$, and it is possible to integrate again from $s = 0$, $u = u_o$ to $s = s_o$, $u = u_1$. Then:

$$u_o - u_1 = \frac{q\, s_o^2}{2\lambda} = \Delta t \quad \ldots\ldots\ldots \quad (3)$$

where $\Delta t$ is the temperature difference from the median plane to the cooling walls.

## 2. Inner-catalyst Converter

Nomenclature (refer to fig. 8)

$r_o$ = tube inside radius (m).

$r$ = radial distance out to any point in the catalyst bed. (Other symbols as in case 1.)

For an infinite cylinder with circular symmetry, in the steady state the temperature distribution is given by

$$\lambda \left[ \frac{1}{r} \frac{du}{dr} + \frac{d^2u}{dr^2} \right] + q = 0 \quad \ldots\ldots \quad (4)$$



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Rearranging:

$$\frac{du}{dr} + r\frac{d^2u}{dr^2} = \frac{-qr}{\lambda} \quad \ldots\ldots\ldots (5)$$

Integrating:

$$r\frac{du}{dr} = \frac{-qr^2}{2\lambda} + C_1 \quad \ldots\ldots\ldots\ldots\ldots (6)$$

However, at $r = 0, \frac{du}{dr} = 0$, and $C_1 = 0$, so that:

$$\frac{du}{dr} = \frac{-qr}{2\lambda} \quad \ldots\ldots\ldots\ldots\ldots\ldots (7)$$

Integrating again from $r = 0$, $u = u_o$ to $r = r_o$, $u = u_1$:

$$\Delta t = u_o - u_1 = \frac{q\, r_o^2}{4\lambda} \quad \ldots\ldots\ldots\ldots (8)$$

where $\Delta t$ is the temperature drop from the center of the tube to the cooling wall.

### 3. Outer-catalyst Converter

Referring to figure 9 and considering that the tubes are centered in an equilateral triangular pattern, it is evident that, for this case, the catalyst bed may be subdivided into regular hexagonal volumes with a central hole (the cooling tube). The exact general solution for steady-state temperature distribution in such a unit volume is not available at present. As an approximation, the hexagonal boundary may be replaced with an average circular one whose radius is given by:

$$r_1 = \frac{1}{2}\left[\frac{\chi}{2} + \frac{\chi}{\sqrt{3}}\right] = 0.5386\,\chi$$

where $\chi$ is the center-to-center tube spacing. Figure 10 shows this equivalent circular catalyst arrangement.

Nomenclature:

$r_o$ = outer radius of cooling tubes (m).

$r_1$ = "average" radius of hexagon (m).

(Other symbols as before.)



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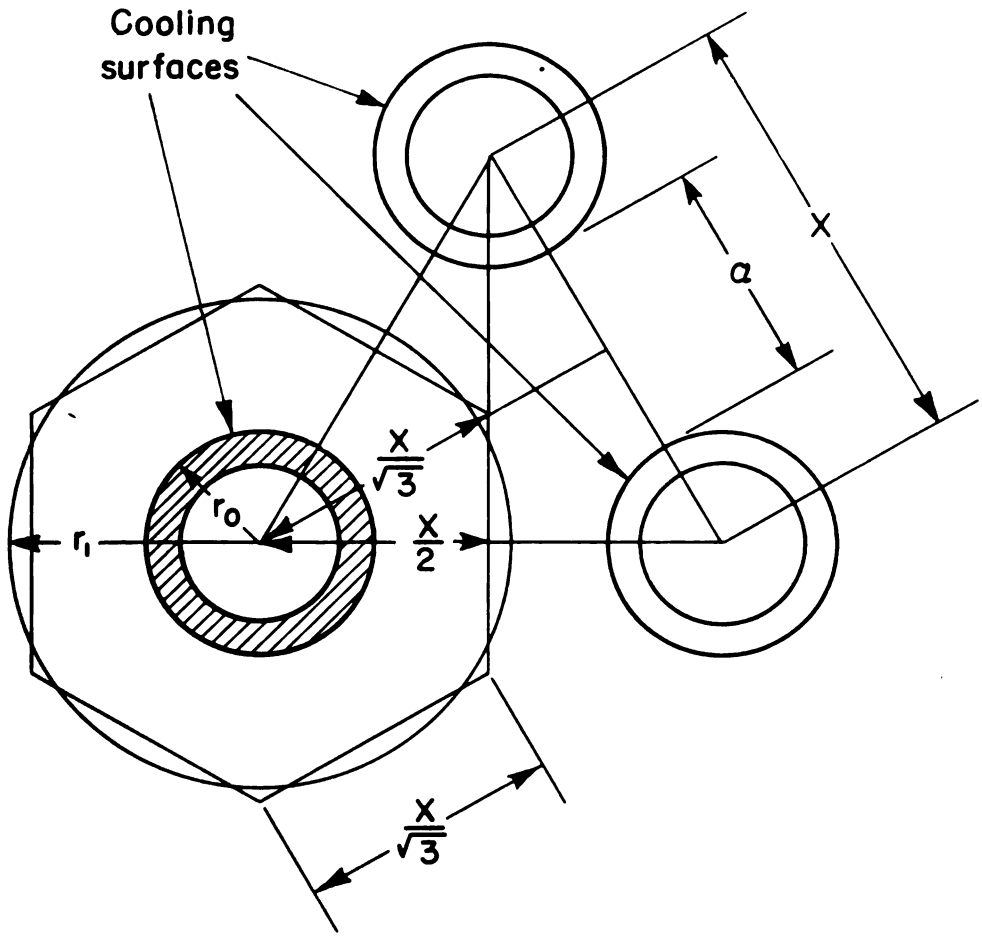


Figure 9. - Tube converter in which the catalyst surrounds the tube through which flows the cooling medium (outer-catalyst converter).

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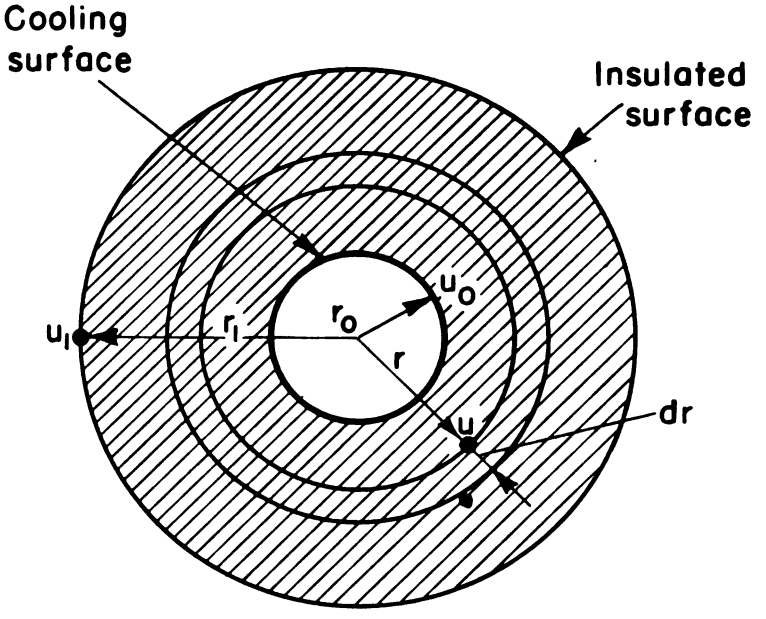


Figure 10. - Converter with a catalyst layer in the form of a circular ring.

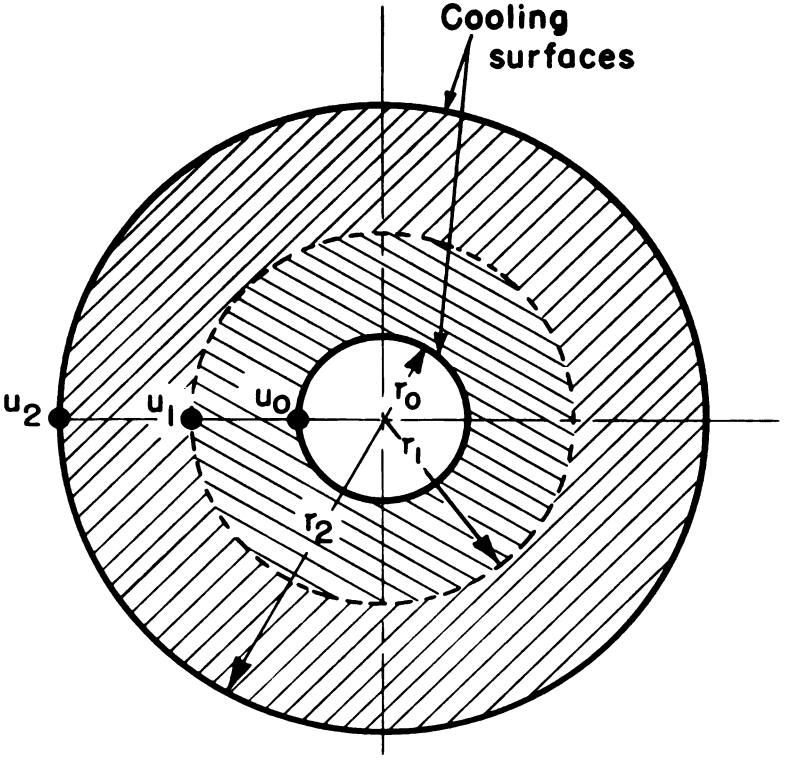


Figure 11. - Section through an element of the annular-volume converter.

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Returning to equation (6) above, and changing signs:

$$-r \frac{du}{dr} = \frac{qr^2}{2\lambda} + C_1 \quad \ldots\ldots\ldots\ldots (9)$$

However, at $r = r_1$, $\frac{du}{dr} = 0$ (no heat flow across hexagon boundary, by virtue of symmetry), hence

$$C_1 = \frac{qr_1^2}{2\lambda}$$

so that $$r \frac{du}{dr} = \frac{qr^2}{2\lambda} - \frac{qr_1^2}{2\lambda} \quad \ldots\ldots\ldots\ldots\ldots\ldots (10)$$

Rearranging and integrating from $r = r_o$, $u = u_o$ to $r = r_1$, $u = u_1$:

$$\Delta t = u_1 - u_o = \frac{q}{4\lambda} \left[ r_1^2 \left( 2 \ln \frac{r_1}{r_o} - 1 \right) + r_o^2 \right] \ldots\ldots (11)$$

where $\Delta t$ is an approximation to the temperature drop from the hot spots in the catalyst bed (points farthest from the cooling tubes) to the cooling tubes. Equation (11) may be solved by trial to find the average hexagon diameter, $r_1$ (and hence the tube spacing), if the thermal conductivity, cooling-tube radius, and heat-generation rate are known. It differs from equation (3) for the plate converter and equation (8) for the inner-catalyst converter in that the latter may both be solved directly without trial and error methods. Equation (11) is reasonably valid as an approximation, provided the gas flows along (parallel to) the tubes. For cross flow, the exact mathematical analysis is still more difficult, and in discussing this design, equation (11) has to be used as a very crude approximation.

## 4. Annular-volume Converter

Referring to figure 11, it will be noted that the catalyst in an annular section with both inner and outer cooling may be split by a cylindrical surface (of radius $r_1$) across which no heat flows.

Considering the material inside of this cylinder ($r_1 - r_o$), we have directly from case 3 above:

$$\Delta t_i = \frac{q}{4\lambda} \left[ r_1^2 \left( 2 \ln \frac{r_1}{r_o} - 1 \right) + r_o^2 \right] \ldots.(12a)$$

Likewise, returning to equation 10, adjusting signs and integrating from $r = r_1$, $u = u_1$ to $r = r_2$, $u = u_2 = u_o$, gives, for the material in the outer cylinder ($r_2 - r_1$):

$$\Delta t_o = \frac{q}{4\lambda} \left[ r_1^2 \left( 2 \ln \frac{r_1}{r_2} - 1 \right) + r_2^2 \right] \ldots.(12b)$$



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## DISCUSSION

For evaluation of the individual designs, the equations somewhat modified are arranged in the following order:

Plate converter: $$s_o = 1.414\sqrt{\frac{\lambda \cdot \Delta t}{q}} \qquad (3)$$

Inner-catalyst converter: $$r_o = 2\sqrt{\frac{\lambda \cdot \Delta t}{q}} \qquad (8)$$

Outer-catalyst converter: $$r_1^2 \cdot \left( 2 \ln \frac{r_1}{r_o} - 1 \right) + r_o^2 = \frac{4 \cdot \lambda \cdot \Delta t}{q} \qquad (11)$$

Annular-volume converter:

Inside section: $$r_1^2 \cdot \left( 2 \cdot \ln \frac{r_1}{r_o} - 1 \right) + r_o^2 = \frac{4 \cdot \lambda \cdot \Delta t}{q} \qquad (12a)$$

Outside section: $$r_1^2 \cdot \left( 2 \cdot \ln \frac{r_1}{r_2} - 1 \right) + r_2^2 = \frac{4 \cdot \lambda \cdot \Delta t}{q} \qquad (12b)$$

### Comparison of Individual Catalyst Sections

#### Equivalent Catalyst Cross Sections

If one prescribes the same values for $\lambda$, q, and $\Delta t$ in these equations, one obtains equivalent catalyst sections, that is, catalyst beds in which the temperature drops are the same. For assumed values of $\lambda$, q, and $\Delta t$, the distance between cooling surfaces in a plate converter, the diameter of the tubes in the inner-catalyst converter, the width of the catalyst bed, $r_1 - r_o$, of the outer-catalyst converter, and the annular width, $r_2 - r_o$, in the annular-volume converter were calculated and are shown in figure 12. The inside diameter of the tube containing catalyst was selected as the abscissa, and the corresponding dimensions of the other designs were chosen as ordinates. The cooling tube in the outer-catalyst converter and that on the inside in the annular-volume converter were assumed, for the present, to have the same dimensions as the tube in the inner-catalyst converter. As can be seen immediately from a comparison of equations (3) and (8), the ratio of the tube diameter of the inner-catalyst converter to the distance between cooling surfaces of the plate converter was 2/1.41. A straight line is thus obtained in figure 12 for the plate converter, the slope of which is the ratio 1.41/2. The values obtained from equation 11 for double the catalyst space width, $2 (r_1 - r_o)$, in the outer-catalyst converter are somewhat smaller than the corresponding values for the distance, $2 s_o$, between plates in the plate converter, and the curve for these values for the outer-catalyst converter lies under the straight line obtained for the plate converter, as shown in figure 12. The annular width, $r_2 - r_o$, for catalyst situated between two concentric cooling tubes is also shown in this figure. This value varies linearly with



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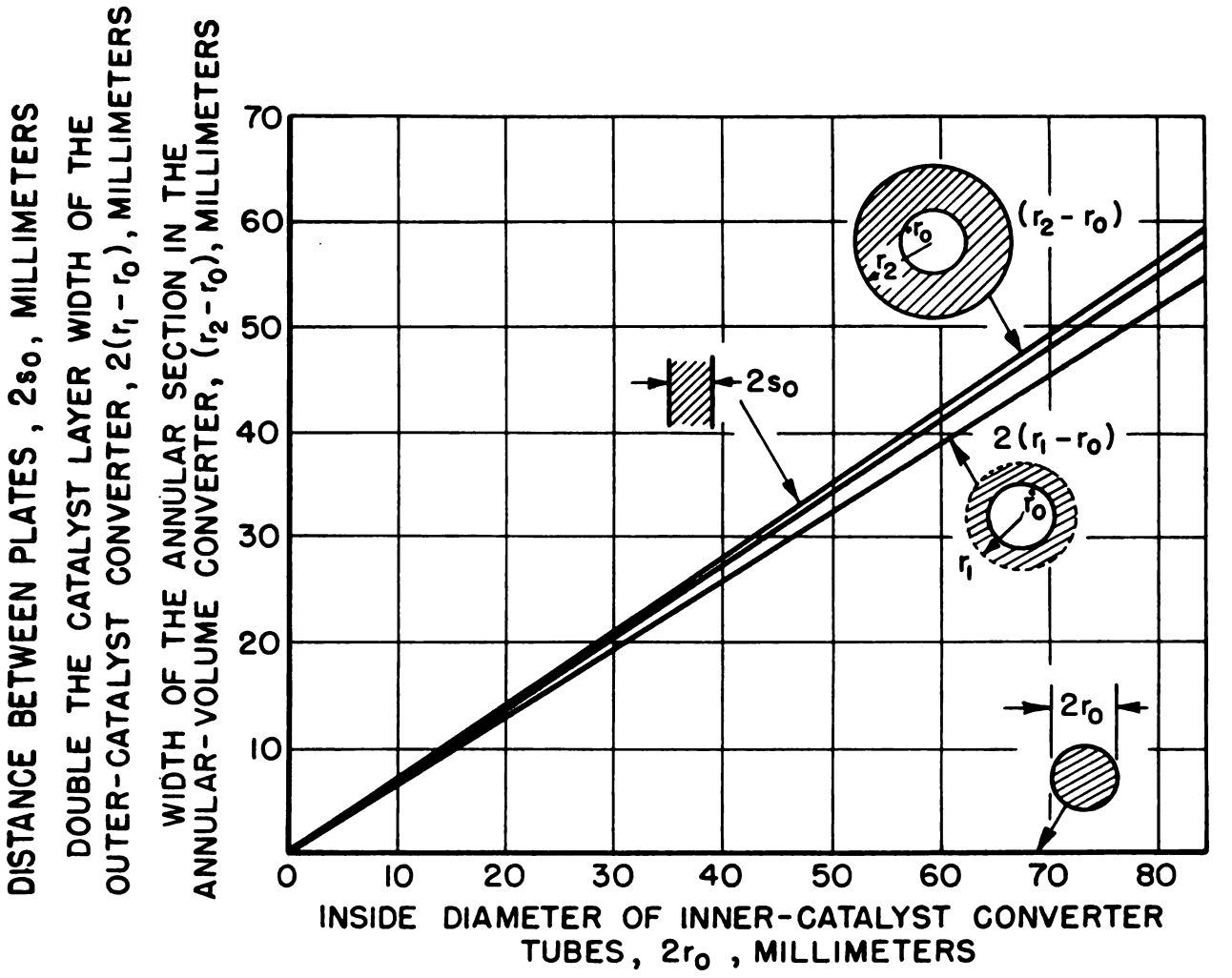


Figure 12. - The inner-catalyst converter element which is equivalent to elements of the plate converter, the outer-catalyst converter, and the annular-volume converter.

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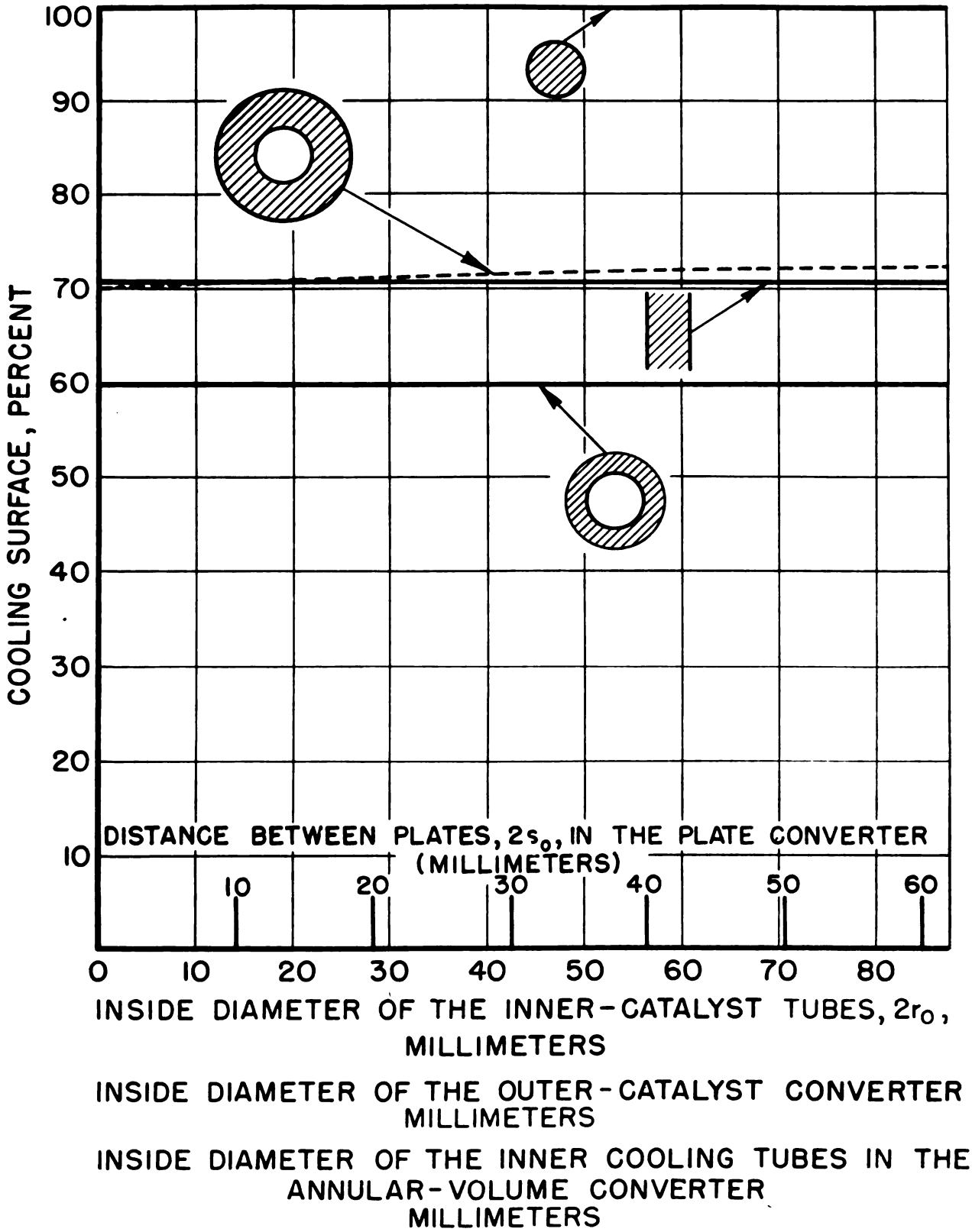


Figure 13. - Comparison of cooling surfaces for various reactor designs. (Cooling surface of the inner-catalyst converter = 100 percent.)

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diameter and differs only slightly from the distance between cooling surfaces in the plate converter.

For example, to obtain equal temperature gradients, it is necessary to use a 51-mm. diameter tube in an inner-catalyst converter, to have 36 mm. between cooling surfaces in a plate-converter, a tube of 51 mm. I.D./57 mm. O.D., for example, and a catalyst section width ($r_1 - r_o$) of 16.7 mm. in an outer-catalyst converter, or an inner cooling tube of 51 mm. I.D./57 mm. O.D. with an annular section width of 35.4 mm. in an annular-volume converter. Ordinary boiler tubes were assumed as a basis for the calculations.

Equivalent Cooling Surfaces

The cooling surfaces per cubic meter of catalyst volume resulting from the dimensions obtained from equations (3), (8), (11), and (12) for the various cross sections were compared with each other, as shown in figure 13. The abscissa was arbitrarily chosen to represent inside diameter of tubes of either the inner-catalyst converter or the outer-catalyst converter; inside diameter of the inner tubes of the annular-volume converter; or the distance between plates in the plate converter. The cooling surface obtained with the inner-catalyst converter was taken as 100 percent on the ordinate scale, and the other values were related to this basis.

It can be seen from figure 13 that, for constant $\Delta t$, q, and $\lambda$, a plate converter requires only about 71 percent of the cooling surface of an inner-catalyst converter and that this is independent of the absolute value of the tube diameters. In the case of the outer-catalyst converter, only about 60 percent of the cooling surface in the inner-catalyst converter is needed, and this is likewise independent of the tube diameter. As the heat-transfer coefficients on the inside of the tubes in the inner-catalyst converter are inferior to those of the outer-catalyst converter, where the outside diameter is used in calculating the cooling surface, it can be seen that (as tubes of the same I.D. and O.D. are used in both converters) the material required for tubes in the outer-catalyst converter is about 60 percent less than that for the inner-catalyst converter. As the tube diameter increases, this advantage decreases (assuming that ordinary boiler tubing is used), as the ratio between outer and inner diameter simultaneously approaches the value 1. The cooling surface of the annular-volume converter amounted to 70-73 percent of that of the inner-catalyst converter and had approximately the same value as that resulting from location of the catalyst between flat cooling plates. The convex inner surface and the concave outer surface of the annular section apparently compensate each other.

## Comparison of Converters

### Cooling-surface Requirements of the Various Designs

Up to this point, all considerations have been based on the assumption of only one path of contact. Consideration of the entire converter may be made on the basis that the plate converter, the inner-catalyst converter, and the annular-volume converter consist of the connection of many individual elements in parallel. In the case of the outer-catalyst converter, however, the



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distance "a" between two cooling tubes (fig. 9) is not the same as the double catalyst-section width 2 ($r_1 - r_o$) calculated from equation 11. As shown in figure 9, the distance between two cooling tubes is

$$x = 2r_o + a$$

and therefore,

$$a = \frac{2r_1}{1.0772} - r_o \qquad \text{..... (13)}$$

The value $r_1$, determined for the outer radius of the annular section from equation 11, must be reduced by the factor 1.0772 in order to obtain the distance, a, between the tubes. Figure 14 shows a comparison of the values for the width of catalyst section ($r_1$), obtained from equation 11, and the values for half the distance between two cooling tubes (a/2), obtained from equation 13, for three cases in which the outer-catalyst converter is equivalent to inner-catalyst converters with tubes of 14, 20, and 50 mm. diameter, respectively. The outer diameter of the cooling tubes was chosen as the abscissa.

The catalyst section width ($r_1 - r_o$) surrounding a single tube increases with increasing cooling-tube diameter, as can be seen in figure 14, and approaches asymtopically the value for a plate converter, which is reached when $r_o = \infty$. Correspondingly, the value, $\frac{a}{2} = \frac{r_1}{1.0772} - r_o$, for half the distance between tubes increases initially with increasing tube diameter but decreases later. The simplifying assumption, that is, that the hexagonal cross section of the catalyst bed surrounding a tube can be effectively considered as a circle, becomes less valid as the cooling-tube diameter increases and the temperature restrictions become more severe; this means that the outer-catalyst converter can be characterized by calculations involving this assumption only when the distance between tubes is small or when the equivalent inner-catalyst converter has a small diameter.

An outer-catalyst converter with tubes 40 mm. O.D. and with heat removal as good as an inner-catalyst converter with 50-mm. tubes must have a distance between cooling tubes of 26.6 mm., whereas, if 10 mm.-diameter cooling tubes are used a distance of 25.2 mm. must be maintained. The corresponding values, under the assumption of a circular catalyst-bed cross section, as obtained from figure 14, amount to 29.8 and 27.8 mm., respectively. As previously noted, the difference between ($r_1 - r_o$) and a/2 becomes smaller with decreasing cooling-tube diameter, and, in view of these considerations, it appears desirable to equip an outer-catalyst converter with the smallest possible cooling tubes.

Figure 15 shows how the width of the annular section depends on the outside diameter of the inner cooling tube for an annular-volume converter in the three cases where the annular-volume converter is equivalent to an inner-catalyst converter of 14, 20, or 50 mm. I.D.

Where the inner cooling-tube diameter is zero, the width of the annular section is identical to the radius of the inner-catalyst converter. From this limiting value, the annular section width increases with increasing cooling-tube diameter, at first rapidly, then more slowly, and becomes approximately



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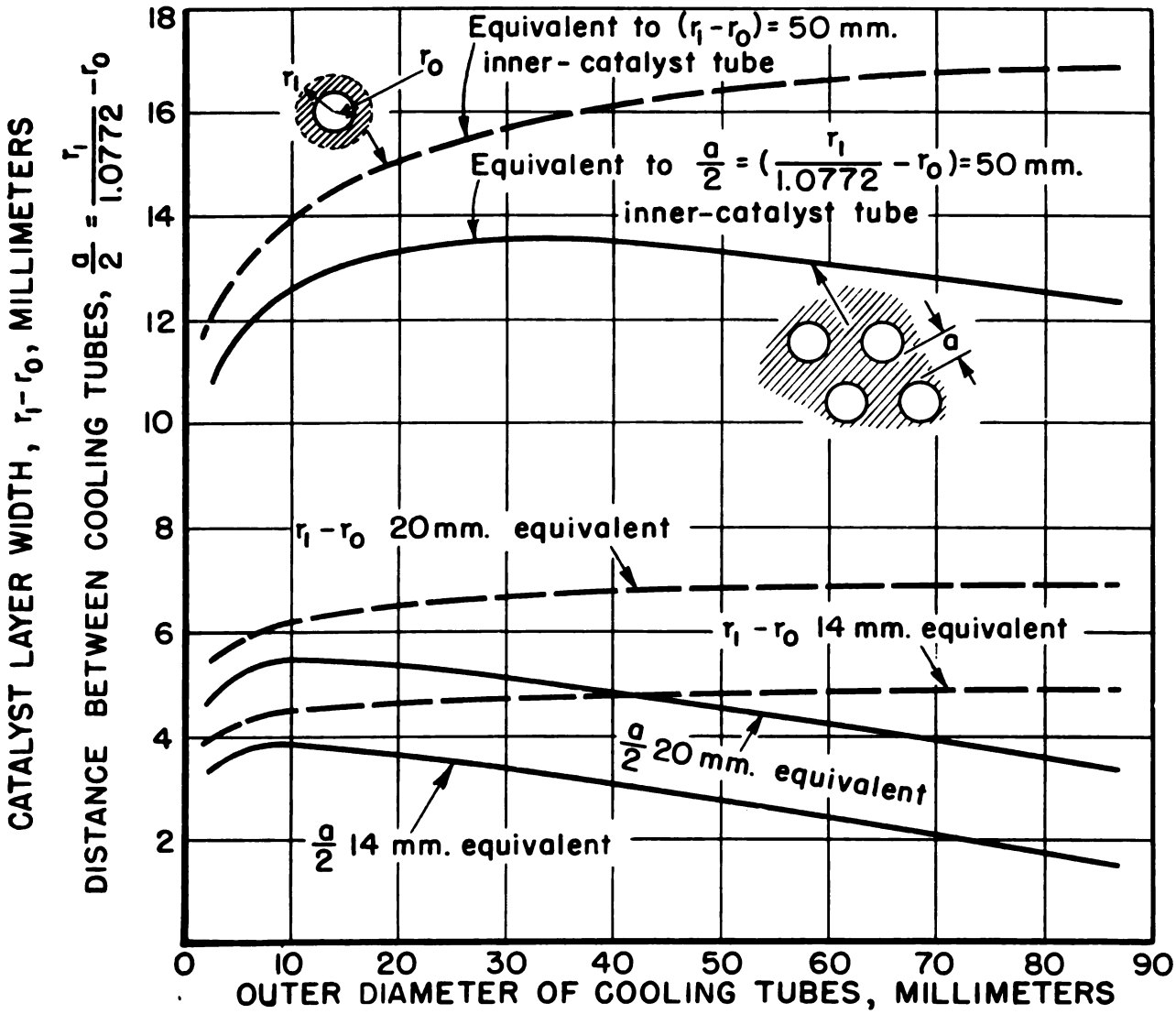


Figure 14. - Dependence of the width of catalyst layer, $r_1-r_0$, in the single tube converter and of half the distance between cooling tubes, $\frac{a}{2}$, in the multiple tube converter with outside catalyst on the outside diameter of the cooling tubes for various inner-catalyst converters of equivalent tube diameters.

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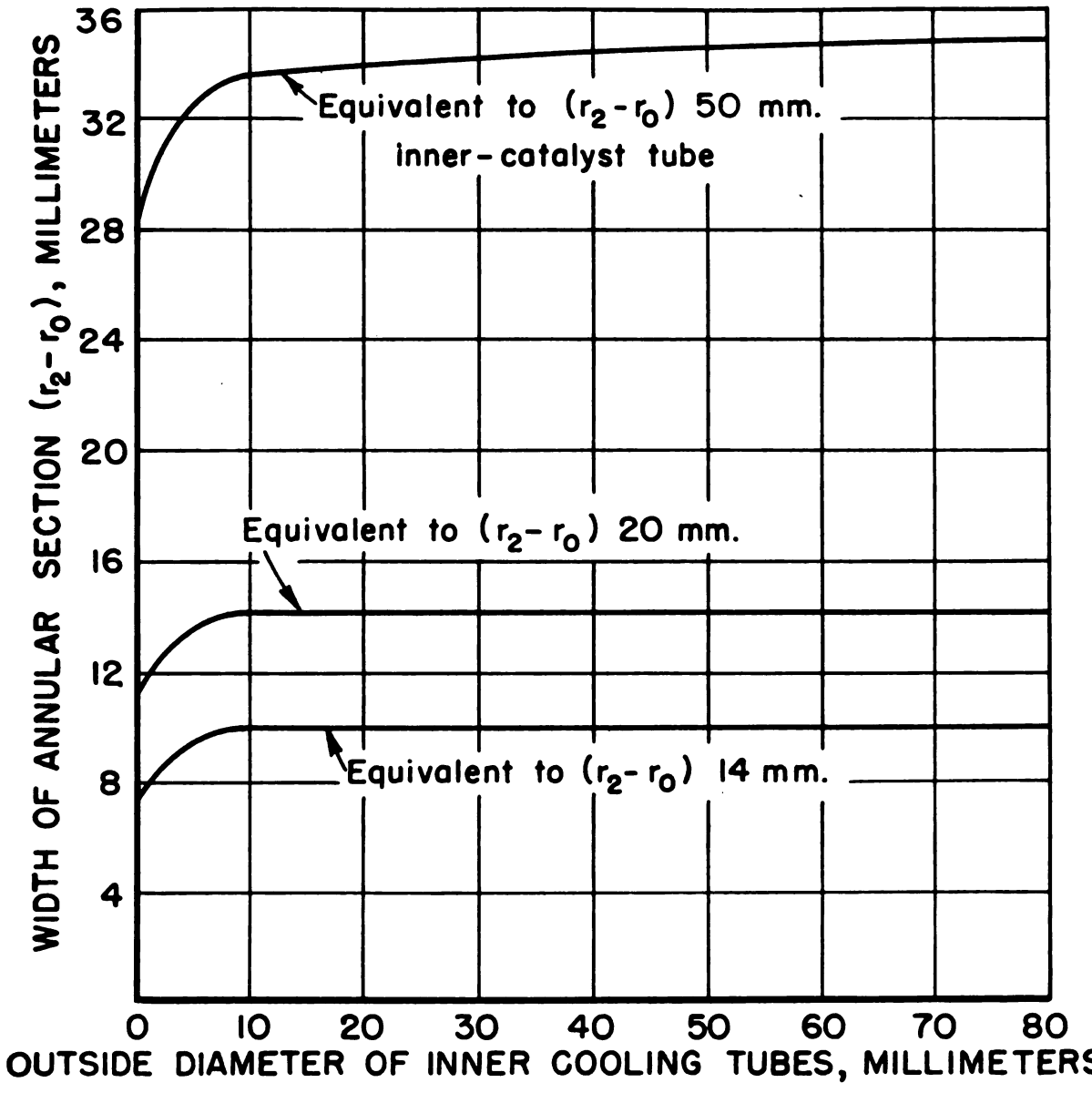


Figure 15. - Dependence of the width of annular section in the annular-volume converter with inner and outer cooling on the outer diameter of the inner cooling tubes.

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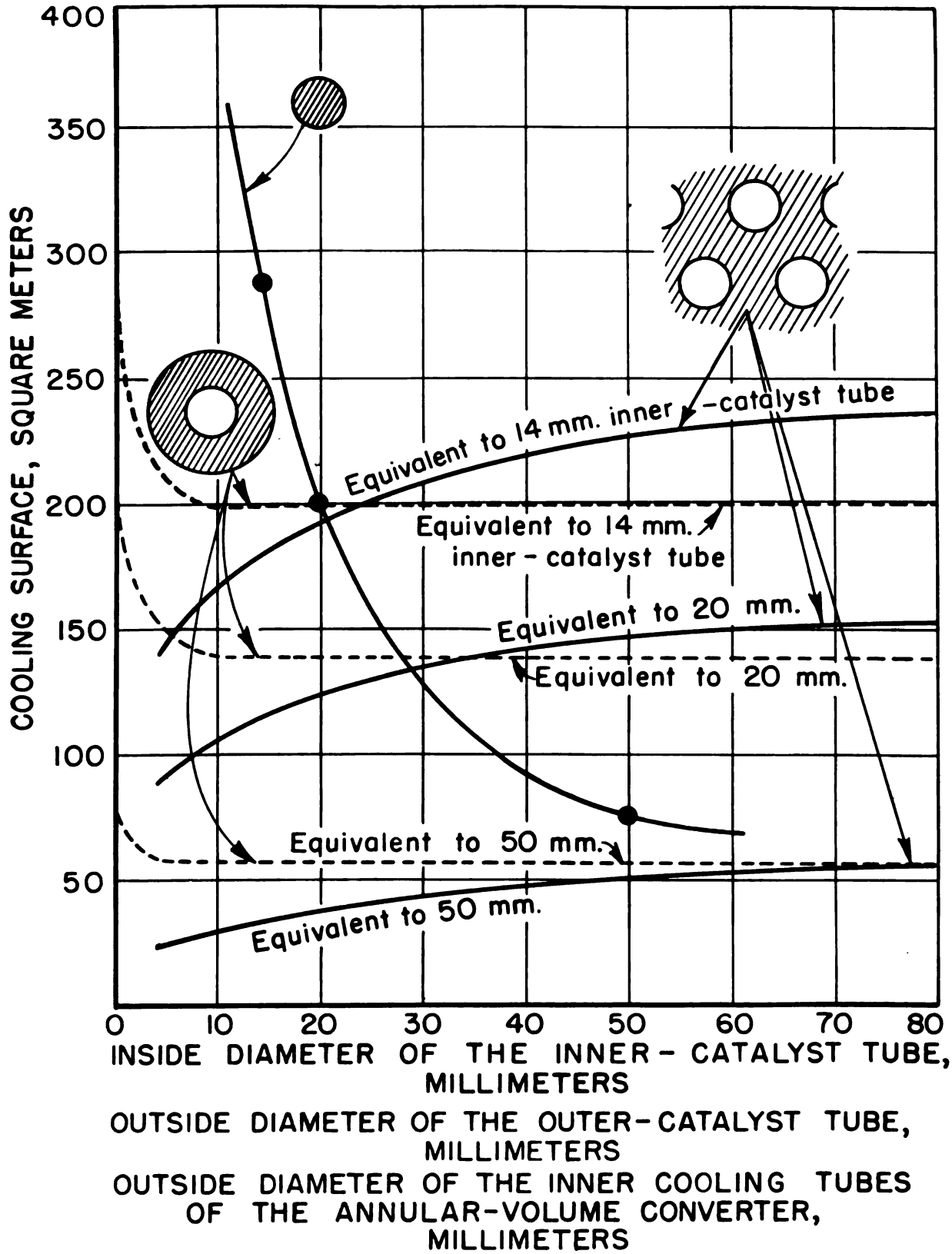


**Figure 16. - Cooling surface per cubic meter of bulk catalyst volume for various types of converters.**

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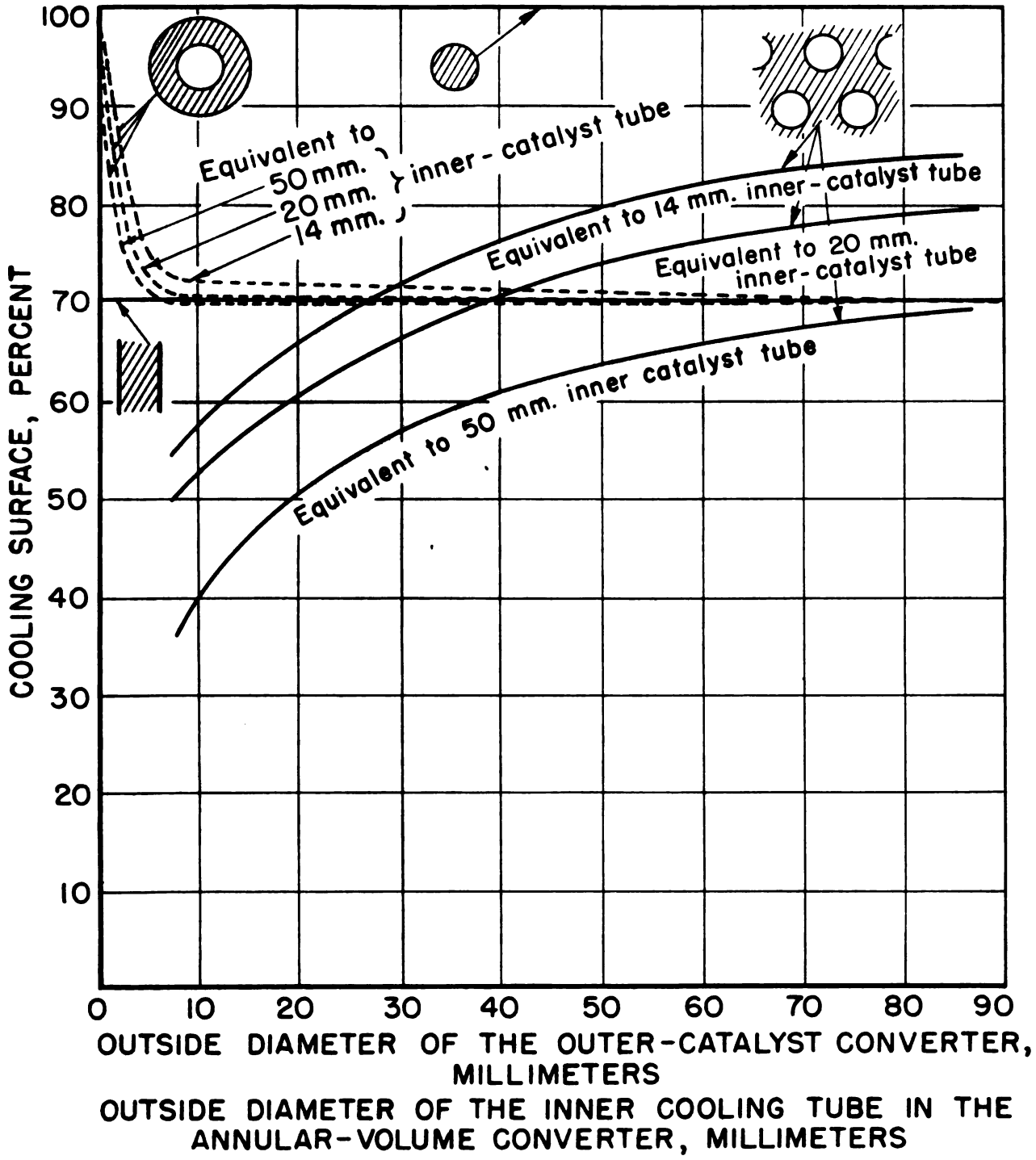


Figure 17. - Comparison of the cooling surfaces of various converter designs. (Cooling surface of the inner-catalyst converter = 100 percent.)

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constant for diameters greater than 10 mm.; its absolute value is about equal to the distance between plates in an equivalent plate converter. The customary calculations for the annular-volume converter can be modified in large-scale operations by using the formulae for a plate converter to calculate the annular width without introducing any significant error, as long as diameters smaller than 10 mm. are avoided for the inner cooling tubes.

The cooling surfaces obtained in 1 cubic meter of catalyst space were ascertained by the dimensions obtained from equations (3) and (8) for plate- and inner-catalyst converters and by the values obtained from figures 14 and 15 for outer-catalyst and annular-volume converters; these surfaces are compared in figure 16. The inside diameter of the inner-catalyst converter and the outside diameter of (1) a cooling tube in the outer-catalyst converter and (2) an inner cooling tube in the annular-volume converter were arbitrarily chosen as the abscissa in figure 16 whereas the cooling surface per cubic meter of catalyst space was assumed as the ordinate.

Inner-catalyst converters of 14, 20, and 50 mm. diameter have cooling surfaces of 286, 200, and 80 $m.^2/m.^3$ of catalyst space, respectively. Three curves for outer-catalyst converters, equivalent, respectively, to the 14, 20, and 50 mm.-diameter inner-catalyst converters, are shown in the same figure; the cooling surface is seen to increase with increasing cooling-tube diameter, but the value never reaches that of the corresponding inner-catalyst converter. If it is desired to construct an outer-catalyst converter equivalent to an inner-catalyst converter of 14 mm. diameter, a cooling surface of 164 $m.^2/m.^3$ catalyst will result by using cooling tubes of 10 mm. diameter, or 208 $m.^2/m.^3$ catalyst by using 30-mm. tubes.

In the annular-volume converter, the proportions are somewhat different. It can be seen from figure 16 that, independent of the diameter of the inner cooling tubes, the cooling surface is maintained virtually constant over a wide range under the assumption that the annular-volume converter is equivalent to an inner-catalyst converter of a given diameter. The values for annular-volume converters equivalent to 14, 20, and 50 mm.-diameter inner-catalyst converters are shown in figure 16. The cooling surfaces of the annular-volume converters are less than those of the inner-catalyst converters, except in the region of very small cooling-tube diameters; below 10 mm. diameter the cooling surface increases rapidly with decreasing diameter and approaches the value for an inner-catalyst converter at $d = 0$. On this basis, it is not desirable to choose inner cooling tubes under 10 mm. diameter for an annular-volume converter. Whereas figure 16 shows the absolute values of the cooling-surface requirements for the various converter designs, figure 17 shows the relative values referred to a basis of 100 percent established for the inner-catalyst converter. The same scale was chosen as the abscissa as that used in figure 16, whereas, as the ordinate, the cooling surface in percent was used.

It is apparent that the cooling-surface requirements of an outer-catalyst converter decrease with decreasing diameter of the cooling tubes regardless of whether it is equivalent to a 14-, 20-, or 50-mm. inner-catalyst converter; this is in contrast to the annular-volume converter. The larger the diameter of the inner-catalyst converter, the greater the percent savings in the cooling surface of the outer-catalyst converter, and this fact is indicated by the corresponding permissible temperature difference in the catalyst bed.



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In the annular-volume converter, the cooling-surface requirements for inner cooling tubes with diameters larger than 10 mm. are about 71 percent of those for the inner-catalyst converter and are almost entirely independent of the size of the inner cooling tubes, as shown in the corresponding temperature distribution. With inner cooling-tube diameters less than 10 mm., the cooling-surface requirements increase until a value of 100 percent is reached at zero.

In the same diagram, the cooling-surface requirements of the plate converter are shown to be a constant ratio of about 71 percent, which is immediately evident from comparison of equations (3) and (8).

Space Utilization of the Various Designs

A further characteristic for the evaluation of a reactor is the efficiency of utilization of the designed volume; the designed volume was defined to include the catalyst, the cooling devices, and the liquid cooling devices, and the liquid cooling medium. The adjacent space, which served to conduct the gas to and from the reaction zone, as well as the space that was eventually required for the construction of a high-pressure jacket was not considered, as the many construction details encountered were determined from theoretical investigations built on a framework of simplifying assumptions.

It was further assumed for the inner-catalyst converter, just as in the annular-volume converter, that the distance between the tubes, which were rolled or welded into the base plates, was three times the wall thickness. In figure 18, the catalyst space (percent of the designed volume) is shown for the various converter designs as a function of (1) the inside diameter of the inner-catalyst converter; (2) the outer diameter of the inner cooling tubes of the annular-volume converter for three cases, equivalent to inner-catalyst converters of 14-, 20-, and 50-mm. inside diameter, and (3) the outer diameter of the cooling tubes in the outer-catalyst converter for three similar cases.

For the inner-catalyst converter, the space utilization increases with increasing tube diameter from 0 to 54 percent at a diameter of 50 mm. By means of other tubing arrangements - for example, by welding the tubes at smaller intervals - improvements could be achieved, particularly with small-diameter tubes. However, increasing the tube diameter would also increase the difficulties of heat removal or steam removal on the side of the cooling medium.

The outer-catalyst converter shows that an increasing percentage of the designed volume is filled with catalyst as the cooling-tube diameter decreases. For a cooling-tube diameter of zero, a value of 100 percent is obtained. Practically, tubes smaller than 10-mm. diameter should not be used, so that this limiting value is never reached. As far as the reactor design is concerned, the less sensitive the reaction is to temperature, the better will be the utilization of space. If the converter is equivalent to an inner-catalyst converter of 50 mm. diameter, a space utilization of 83 percent is obtained with cooling tubes of 20 mm. diameter; whereas with the same cooling-tube diameter, but corresponding only to a 14 mm. inner-catalyst converter, a space utilization of only 51 percent is possible.



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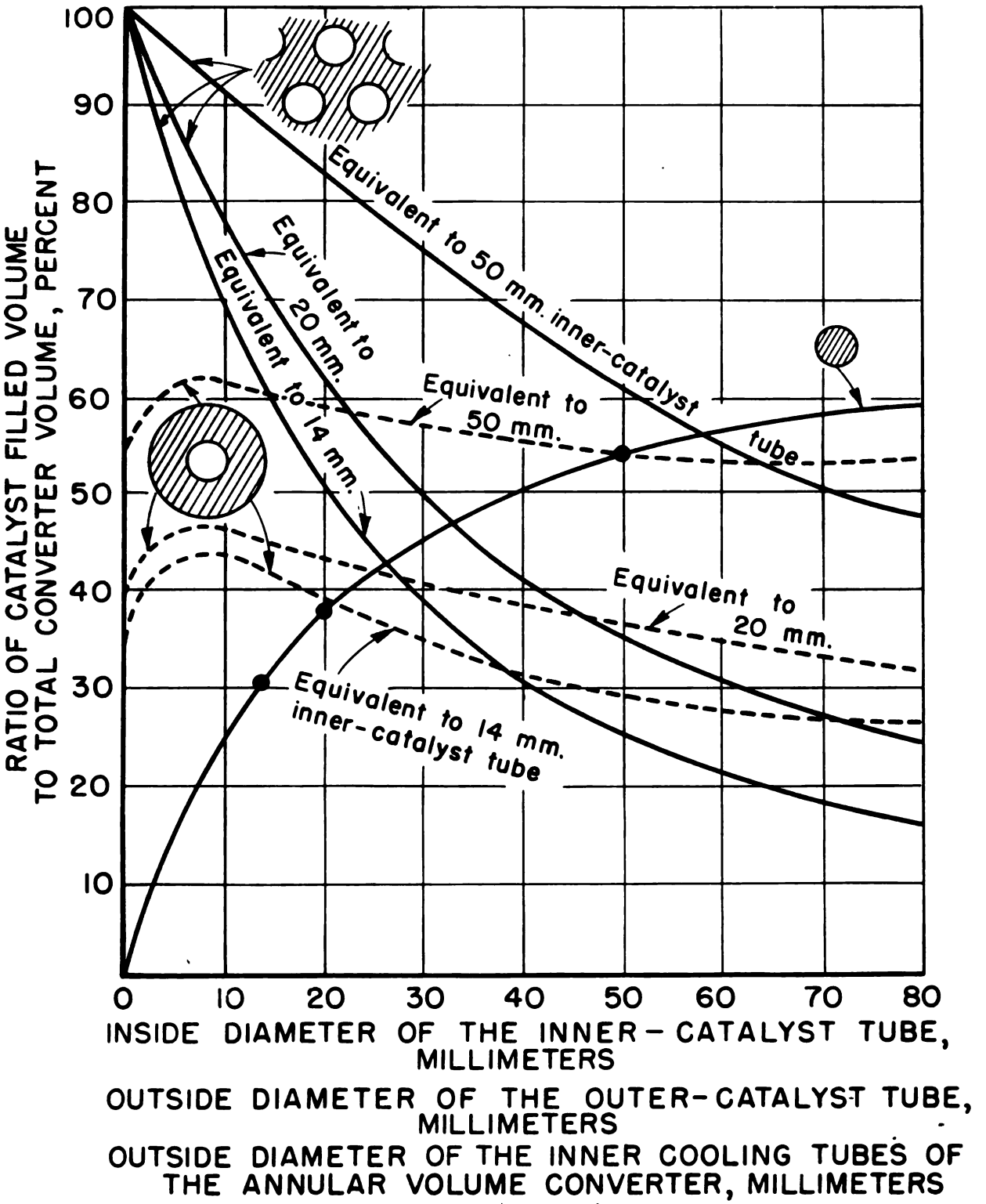


Figure 18. - Space utilization of the various converter designs.

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The curves for the annular-volume converter increase with increasing diameter of the inside cooling tube until a diameter of 10 mm. is reached, at which point they decrease slowly again. The optimum converter design of this type exists at an inner-cooling-tube diameter of 10 mm. At an inner cooling-tube diameter of zero, the space utilization corresponds to that of the inner-catalyst converter. Just as in the other types of converter designs, improved space utilization results with increasing diameter of the equivalent inner-catalyst converter.

## CONCLUSIONS

A method of calculation was developed for various types of converters for reactions that proceed inside narrow temperature limits. With the help of the evolved formulae, the individual types of construction were compared with regard to cooling surface and space utilization. To select a converter of the proper size, it is necessary to consider still other factors that do not appear in the calculations; for example, height of the catalyst bed, linear gas velocity, ease of discharging, construction costs, and maintenance and operating costs.



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